

AD-A129 057

ANALYSIS OF THERMAL CONDUCTIVITY DATA FOR FOURTEEN  
ELEMENTS IN NORMAL AND... (U) THERMOPHYSICAL AND  
ELECTRONIC PROPERTIES INFORMATION ANALYSIS..

1/1

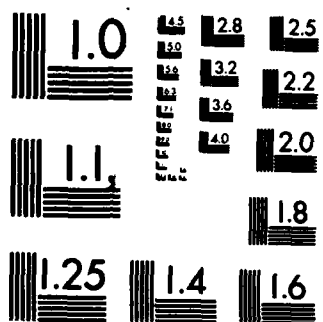
UNCLASSIFIED

J A RAFALOWICZ JUL 74 CINDAS-TPRC-28

F/G 20/3

NL

END
DATE
FILED
DTIC



TPRC

AD A129057

# THERMOPHYSICAL PROPERTIES RESEARCH CENTER

ANALYSIS OF THERMAL CONDUCTIVITY DATA FOR FOURTEEN ELEMENTS  
IN NORMAL AND SUPERCONDUCTING STATES

by  
J. A. Rafalowicz

DTIC  
ELECTE  
JUN 8 1983  
S D

TPRC Report 28

July 1974

This document has been approved  
for public release and sale; its  
distribution is unlimited.

FILE

DTIC FILE COPY

THERMOPHYSICAL PROPERTIES RESEARCH CENTER  
Purdue University  
2595 Yeager Road  
West Lafayette, Indiana 47906

83 06 07 102

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
	AD-A129257	
4. TITLE (and Subtitle)		5. TYPE OF REPORT & PERIOD COVERED
ANALYSIS OF THERMAL CONDUCTIVITY DATA FOR FOURTEEN ELEMENTS IN NORMAL AND SUPERCONDUCTING STATES		State-of-the-Art Report
7. AUTHOR(s)		6. PERFORMING ORG. REPORT NUMBER
J. A. Rafalowicz		TPRC Report 28
9. PERFORMING ORGANIZATION NAME AND ADDRESS		8. CONTRACT OR GRANT NUMBER(s)
Thermophysical and Electronic Properties Information Analysis Center, CINDAS/Purdue Univ., 2595 Yeager Road, W. Lafayette, IN 47906		DSA-900-74-C-2428
11. CONTROLLING OFFICE NAME AND ADDRESS		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
Defense Technical Information Center, Defense Logistics Agency, Attn: DTIC-AI, Cameron Station, Alexandria, VA 22314		
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		12. REPORT DATE
Army Materials and Mechanics Research Center Attn: DRXMR-P Arsenal Street Watertown, MA 02172		July 1974
16. DISTRIBUTION STATEMENT (of this Report)		13. NUMBER OF PAGES
Distribution unlimited		35
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		15. SECURITY CLASS. (of this report)
		Unclassified
18. SUPPLEMENTARY NOTES		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
TEPIAC Publication (DTIC Source Code 413571); Microfiche copies available from DTIC		N/A
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) *Thermal conductivity--- *Superconductivity---*Low temperature data---*Metals---*Elements---Aluminum--- Zinc---Cadmium---Gallium---Rhenium---Lead---Indium---Tin---Mercury---Thallium--- Niobium---Tantalum---Vanadium---Lanthanum		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Using thermal conductivity data from TPRC's data bank for fourteen metallic elements in both normal and superconducting states, the super- conducting transition temperature, the thermal conductivity at the transition temperature, and the purity of the samples have been estimated by graphical methods. Two basic relationships correlating the thermal conductivity at the transition temperature with the sample purity and correlating the transition temperature with the thermal conductivity at the transition temperature have been established for twelve of the fourteen elements investigated.		

DD FORM 1 JAN 73 1473

EDITION OF 1 NOV 65 IS OBSOLETE

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

**ANALYSIS OF THERMAL CONDUCTIVITY DATA FOR FOURTEEN ELEMENTS  
IN NORMAL AND SUPERCONDUCTING STATES**

by  
**J. A. Rafalowicz**

**TPRC Report 28**

**July 1974**

Accession For	
NTIS GRA&I	<input checked="" type="checkbox"/>
DTIC TAB	<input checked="" type="checkbox"/>
Unannounced	<input checked="" type="checkbox"/>
Justification	
By _____	
Distribution/	
Availability Codes	
Dist	Avail and/or Special
<input checked="" type="checkbox"/>	



**THERMOPHYSICAL PROPERTIES RESEARCH CENTER**  
Purdue University  
2595 Yeager Road  
West Lafayette, Indiana 47906

## FOREWORD

This work constitutes a preliminary study conducted at the Thermophysical Properties Research Center made possible by the five-month residency of Dr. Jerzy A. Rafalowicz as a guest worker and as a United Nations Fellow. Dr. Rafalowicz is affiliated with the Institute for Low Temperature and Structure Research, Polish Academy of Sciences, 95 Prochnik Street, Wroclaw, Poland.

The subject of superconductivity is of great current interest both from scientific as well as technological considerations. Hence, it was most timely to have had an opportunity to review this problem in some detail as has been done in this study. Unfortunately, the limited time available would not allow the pursual of this study in greater depth. Nevertheless, many interesting behavioural characteristics of the thermal conductivity of metals in the superconducting state have been uncovered which certainly deserve follow-through study in the years ahead.

In passing, it is perhaps worthwhile to note that such a study could have been carried out and completed in such a short time period only at a location such as TPRC where comprehensive data banks are available to provide information on demand for a large number of thermophysical properties for a wide spectrum of substances and materials.

## SUMMARY

Using thermal conductivity data from TPRC's data bank [1] \* for fourteen metallic elements in both normal and superconducting states, the superconducting transition temperature, the thermal conductivity at the transition temperature, and the purity of the samples (as expressed by the impurity parameter  $\beta$ ) have been estimated by graphical methods. Two basic relationships correlating the thermal conductivity at the transition temperature with the sample purity (equation 2) and correlating the transition temperature with the thermal conductivity at the transition temperature (equation 6) have been established for twelve of the fourteen elements investigated.

By using the low-temperature thermal conductivity equation (equation 1), the thermal conductivity maximum and its corresponding temperature have been calculated and relations of these quantities to the superconducting transition temperature, to the thermal conductivity at the transition temperature, and to the purity of the samples have been investigated.

The dependence of reduced thermal conductivity on the reduced temperature, for samples of different purity, has been studied and qualitative comparison with theoretical predictions has been made.

Some additional relationships on the dependence of transition temperature and impurity parameter on the sample purity as expressed by the ratio of electrical resistivity, and between the transition temperatures determined from thermal conductivity data and those determined from electrical measurements have been presented.

---

\* Numbers in brackets refer to references at the end of report.

## CONTENTS

	Page
Foreword . . . . .	ii
Summary . . . . .	iii
Notation . . . . .	v
I. Introduction . . . . .	1
II. Relationships between Transition Temperature, Thermal Conductivity at the Transition Temperature, and Sample Purity . . . . .	1
III. Relationships between Thermal Conductivity at the Transition Temperature and Thermal Conductivity Maximum and between Thermal Conductivity Maximum, It Corresponding Temperature, and Transition Temperature . . . . .	4
IV. Relationships between Reduced Thermal Conductivity and Reduced Temperature . . . . .	6
V. Some Additional Observations . . . . .	8
VI. Discussions and Conclusions . . . . .	8
VII. Acknowledgments . . . . .	10
VIII. References . . . . .	10



## NOTATION

$A$	Coefficient, equation (2)
$B$	Coefficient, equation (5)
$C$	Coefficient, equation (7)
$D$	Coefficient, equation (7)
$E$	Coefficient, equation (11)
$k$	Thermal conductivity
$k_c$	Thermal conductivity at the superconducting transition temperature
$k_e$	Electronic thermal conductivity
$k_m$	Thermal conductivity maximum
$k_s$	Thermal conductivity for the superconducting state
$m$	Constant, equation (1')
$n$	Constant, equation (1)
$T$	Temperature
$T_c$	Superconducting transition temperature determined from thermal conductivity data
$T'_c$	Superconducting transition temperature determined from electrical measurement
$T_m$	Temperature of thermal conductivity maximum
$W_c$	Thermal resistivity at the superconducting transition temperature
$W_m$	Thermal resistivity at the temperature of thermal conductivity maximum
$W_o$	Residual electronic thermal resistivity
$\alpha'$	Coefficient, equation (1)
$\alpha''$	Coefficient, equation (1')
$\beta$	Impurity parameter, equation (1)
$\rho_o$	Residual electrical resistivity
$\rho_{273}$	Electrical resistivity at 273 K

## I INTRODUCTION

Of the twenty-five known superconducting elements, experimental thermal conductivity data for the superconducting state are available for only fourteen elements and no data are known to exist for the remaining eleven. The fourteen superconducting elements for which data are available may be divided into three groups.

Aluminum, zinc, cadmium, gallium, and rhenium belong to the first group of superconductors whose superconducting transition temperatures are lower than the temperatures of their respective thermal conductivity maxima. Around the transition temperature, the heat flux is therefore scattered mainly by chemical impurities and lattice imperfections.

Lead, indium, tin, mercury, and thallium belong to the second group of superconductors whose transition temperatures are higher than the temperatures of their respective thermal conductivity maxima. Around the transition temperature, the heat flux is therefore scattered mainly by lattice vibrations (phonons). (how important scattering!)

Niobium, tantalum, vanadium, and lanthanum belong to the third group of superconductors whose transition temperatures are located around the temperatures of their respective thermal conductivity maxima. Around the transition temperature, the heat flux is therefore scattered partly by chemical impurities and lattice imperfections and partly by phonons.

The microscopic theory of thermal conductivity of superconductors is well developed for the first group of superconductors cited above. The theory is moderately developed for the second group of superconductors, and poorly developed for the third group of superconductors.

The available thermal conductivity data for these fourteen superconducting elements in normal and superconducting states have been analyzed and the main aim of this investigation is to find some general relationships and explanations on the basis of present theory.

The results obtained from the data analysis for the fourteen superconducting elements are presented below.

## II. RELATIONSHIPS BETWEEN TRANSITION TEMPERATURE, THERMAL CONDUCTIVITY AT THE TRANSITION TEMPERATURE, AND SAMPLE PURITY

Figure 1 presents the thermal conductivity data for indium, selected as being typical of the dependence of thermal conductivity on temperature in the low temperature

region for a metal in normal and superconducting states. For the purer samples of indium the transition temperatures are higher than the temperatures of thermal conductivity maxima, but for the impure samples the transition temperatures are lower than the temperatures of thermal conductivity maxima. The value of transition temperature for each sample has been obtained graphically as the temperature at which the thermal conductivity curves for the normal and superconducting states intersect and is referred to as  $T_c$ . The value of thermal conductivity at the transition temperature,  $k_c$ , has been read from the graph. Using relations developed at TPRC [2] a thermal conductivity curve in the low temperature region has been drawn for each sample in the normal state as a parallel curve to the TPRC recommended curve, which is only for a sample in the normal state having residual electrical resistivity,  $\rho_0$ , of  $0.00059 \mu\Omega \text{ cm}$ . In this way we have obtained by a graphical method the values of the parameter  $\beta$  contained in the low-temperature thermal conductivity equation [2]:

$$k \approx k_e = (\alpha' T^n + \beta T^{-1})^{-1} \quad (1)$$

where

$$\alpha' = \alpha' \left( \frac{\beta}{n\alpha'} \right)^{(m-n)/(m+1)} \quad (1')$$

$\beta$  is the impurity parameter, and  $\alpha'$ ,  $m$ , and  $n$  are constants for a given metal. From the basic graph of the thermal conductivity dependence on temperature we have thus obtained the values of transition temperatures, thermal conductivity values at transition temperatures, and the  $\beta$  values that are related to the purity of the samples.

Values of transition temperatures,  $T_c$ , and the thermal conductivity at the transition temperatures,  $k_c$ , for samples of different purity for all 14 superconducting metals investigated are collected in Table I.

Figure 2 shows the thermal conductivity at the transition temperature as a function of the impurity parameter,  $\beta$ , for all 14 superconducting elements investigated. It is evident that in the log-log presentations the dependence is linear, and for 12 of the 14 elements the dependence is similar so that a general equation (valid for 12 elements) can be derived which has the form:

$$k_c = A \beta^{-1} \quad \text{For high } \beta, \text{ where } T_c \text{ is low.} \quad (2)$$

where  $A$  is a coefficient of proportionality.

From Figure 2, values of the coefficient A of equation (2) for each element have been estimated and these values are given in Table II.

For indium and mercury the values of the coefficient A were not estimated because the  $k_c$  values of these two elements do not follow the general form of equation (2). However, the dependence of thermal conductivity at the transition temperature on the purity of the samples for indium and mercury can be described by the following equations:

$$\text{In: } k_c = (3.15 \pm 0.05) \beta^{-(0.85 \pm 0.02)} \quad (3)$$

$$\text{Hg: } k_c = (1.0 \pm 0.01) \beta^{-(0.14 \pm 0.01)} \quad (4)$$

Figure 3 shows the dependence of the coefficient A of equation (2) on the transition temperature for 12 elements. The intervals of transition temperatures used are those in which the thermal resistivity at transition temperature is a linear function of transition temperature. From the linear dependence shown in Figure 3 we obtained the equation:

$$A = BT_c \quad (5)$$

where the coefficient B was found to be unity with an accuracy of within a few percent.

By substituting equation (5) into equation (2) we obtain:

$$T_c = \beta k_c \quad (6)$$

Equations (2) and (6) represent two important relations valid for 12 of the 14 superconducting elements investigated. Only indium and mercury do not follow the two equations.

Figure 4 shows a typical dependence of thermal resistivity at transition temperature on the transition temperature. This is for the case of tin. As shown in Figure 4, a linear dependence of thermal resistivity at transition temperature on the transition temperature was noted for impure samples. For the purest samples, however, this linear dependence does not hold. This is true for all the elements investigated. Hence, we may write the following equation for the thermal resistivity at transition temperature:

$$\frac{1}{k_c} = W_c = C - DT_c \quad (7)$$

which is valid for the cases of impure samples for all the 14 superconducting elements investigated. The values of the coefficients C and D for different elements are listed in Table III.

One must remember that equation (7) is more accurate over small temperature ranges than equation (2) which holds to a lesser degree of accuracy over larger temperature ranges.

Figure 5 shows the dependence of transition temperature of lead on the purity of the samples, which is typical for all the elements investigated. It is evident that the transition temperature increases with increasing sample purity.

Substituting  $T_c$  from equation (6) into equation (7) yields the relation:

$$k_c = \frac{C}{2D\beta} \left( 1 + \sqrt{1 - \frac{4D\beta}{C^2}} \right) \quad (8)$$

Substituting  $k_c$  from equation (6) into equation (8) yields the relation:

$$T_c = \frac{C}{D} \left( 1 + \sqrt{1 - \frac{4D\beta}{C^2}} \right) \quad (9)$$

It is easy to see that for  $4D\beta/C^2 \ll 1$  equation (8) reduces to equation (2) and equation (9) to equation (6). Substituting  $k_c$  from equation (2) into equation (7) we may obtain:

$$T_c = \frac{C}{D} - \frac{\beta}{AD} \quad (10)$$

Equations (2, 6-10) give values with different degrees of accuracy.

The approximate ranges of the coefficient  $\beta$  and of transition temperatures  $T_c$  for which equations (2, 6-10) are valid are listed in Table IV. These ranges have been estimated from the graphs drawn for each element.

### III. RELATIONSHIPS BETWEEN THERMAL CONDUCTIVITY AT THE TRANSITION TEMPERATURE AND THERMAL CONDUCTIVITY MAXIMUM AND BETWEEN THERMAL CONDUCTIVITY MAXIMUM, ITS CORRESPONDING TEMPERATURE, AND TRANSITION TEMPERATURE

At low temperatures the thermal conductivity has a maximum value  $k_m$  at a corresponding temperature  $T_m$ . The values of  $k_m$  and  $T_m$  for samples of different purity of each element investigated have been calculated from relations developed earlier [2] using values of the impurity parameter  $\beta$  determined graphically from thermal conductivity plots and are presented in Table V. For the superconducting elements tin, mercury, vanadium, and lanthanum no recommended equations were available at TPRC.

Figure 6 shows the thermal resistivity at the transition temperature as a function of temperature of thermal conductivity maximum for 10 superconducting elements. It is evident that a linear dependence exists in the logarithmic scale utilized. Six of the ten elements investigated have a similar dependence and the general equation determined graphically has the form:

$$W_c = E T_m^{(4.4 \pm 0.2)} \quad (11)$$

where E is the coefficient of proportionality. Equation (11) is valid for aluminum, zinc, gallium, rhenium, lead, and thallium. The values of E for the above mentioned six elements are listed in Table VI. The numerical values of W and  $T_m$  plotted in Figure 6 may be found in the second column of Tables I and V, respectively. For the remaining four superconducting elements the dependence of thermal resistivity at the transition temperature on the temperature of thermal conductivity maximum has the forms given by equations (12) through (15).

$$\text{Cadmium: } W_c = (2.8 \pm 0.1) 10^{-5} T_m^{(6.2 \pm 0.1)} \quad (12)$$

$$\text{Indium: } W_c = (6.6 \pm 0.2) 10^{-4} T_m^{(3.4 \pm 0.1)} \quad (13)$$

$$\text{Niobium: } W_c = (8.5 \pm 0.5) 10^{-5} T_m^{(3.2 \pm 0.1)} \quad (14)$$

$$\text{Tantalum: } W_c = (2.05 \pm 0.05) 10^{-5} T_m^{(3.9 \pm 0.1)} \quad (15)$$

It follows from equation (1) and the expression for  $\alpha'$  that

$$T_m = \left( \frac{\beta}{n \alpha''} \right)^{\frac{1}{m+1}} \quad (16)$$

Solving for  $\beta$  and substituting into equation (2) we obtain

$$W_c = A^{-1} n \alpha'' T_m^{m+1} \quad (17)$$

Equation (17) is equivalent to equation (11). Based on the experimental evidence it is clear that the  $W_c$  vs.  $T_m$  curve for Cd is not parallel to those of the elements from the first group because of the large value of the exponent m for Cd.

If the value of  $\beta$  derived from equation (16) is substituted into equation (10) we obtain:

$$T_c = \frac{C}{D} - \frac{n \alpha''}{A D} T_m^{m+1} \quad (18)$$

Figure 7 shows the dependence of transition temperature on temperature of thermal conductivity maximum for the five superconducting elements belonging to the first group.

If equation (1) is used as an expression for  $k_m$  at temperature  $T_m$  and  $T_m$  is substituted from equation (18), one obtains the following equation:

$$W_m = \frac{1}{k_m} = \alpha'' \left[ \frac{A D}{n \alpha''} \left( \frac{C}{D} - T_c \right) \right]^{\frac{m}{m+1}} + n \alpha'' \left[ \frac{A D}{n \alpha''} \left( \frac{C}{D} - T_c \right) \right]^{\frac{m}{m+1}} \quad (19)$$

The first term of equation (19) is predominant for  $T_c$  near and higher than  $T_m$  and the second term is predominant for  $T_c$  less than  $T_m$ . Figure 8 shows the thermal resistivity at the temperature of thermal conductivity maximum versus the transition temperature for the superconducting elements of the first group. One observes a linear dependence in the logarithmic plot which is in agreement with equation (19).

Again, if equation (1) is used as an expression for  $k_m$  at temperature  $T_m$  and values of  $T_m$  are derived for both the cases where  $T_c$  is lower and higher than  $T_m$  and next these two values of  $T_m$  are substituted into equation (17), one obtains:

$$W_c \cong A^{-1} n^{-\frac{1}{m}} \alpha''^{-\frac{1}{m}} W_m^{\frac{m+1}{m}} \quad (\text{if } T_c < T_m) \quad (20)$$

$$W_c \cong A^{-1} n \alpha''^{-\frac{1}{m}} W_m^{\frac{m+1}{m}} \quad (\text{if } T_c \geq T_m) \quad (21)$$

Figure 9 shows the thermal resistivity at transition temperature versus the thermal resistivity at the temperature of thermal conductivity maximum for the superconducting elements of the first group. Linear dependence in logarithmic plot is obtained, which is in qualitative agreement with equation (20).

#### IV. RELATIONSHIPS BETWEEN REDUCED THERMAL CONDUCTIVITY AND REDUCED TEMPERATURE

According to the phenomenological and microscopic theories of superconductivity the dependence of reduced thermal conductivity (the thermal conductivity at a given temperature for superconducting state,  $k_s$ , divided by the thermal conductivity at the transition temperature) on reduced temperature (the actual temperature divided by the transition temperature) is a universal function for all superconductors [3]. The characteristic of this universal function is different for different groups of superconductors

depending upon whether the transition temperature is higher, lower, or near the temperature of thermal conductivity maximum. Many studies have been made to compare the theory with experimental data. Hence in this brief study an effort is made only to discover the frequency of possible systematic regularities for the three groups of superconducting elements investigated.

In Figure 10 is shown a typical dependence of reduced thermal conductivity on reduced temperature for aluminum samples of different purity. Aluminum is representative of the superconducting elements whose transition temperatures are lower than the temperature of thermal conductivity maxima. The transition temperature for each sample has been estimated from thermal conductivity data. This method is here suggested as an improvement on the procedure used in the investigations where reduced temperature was based on the transition temperature obtained from electrical measurements. Figure 10 presents the family of curves with the purity of the samples as a parameter. Upper curves correspond to the purer samples. On the same graph has been drawn the theoretical curve obtained from the BRT theory for aluminum [4]. The best agreement of theory with experiment is for the samples of Al of average purity. The curves for the purest of samples are above the theoretical curve and the curve for a very impure sample is below the theoretical curve.

Lead is representative of the superconducting elements whose transition temperatures are higher than the temperatures of thermal conductivity maxima. Figure 11 shows the typical dependence of reduced thermal conductivity on reduced temperature for the elements of this group. The behaviour of lead is shown as an example. It is seen that the data for the most pure samples of lead ( $\beta = 0.172$  and  $0.223$ ) lie close to the curve from the so-called strong coupling theory [5] only in the range of reduced temperature  $1.0-0.965$ . In much better agreement with the data for the above mentioned samples is the theory of Gupta and Verma [6]. The data for an impure sample of lead ( $\beta = 3.60$ ) lie close to the curve from the Tewordt theory for Pb [5] in the range of reduced temperature  $1.0-0.84$ . For comparison, the theoretical curves from the BCS theory for Pb [7] and the general BCS theory [7] have been plotted.

Niobium is representative of the superconducting elements whose transition temperatures are near the temperatures of thermal conductivity maxima. Figure 12 shows the dependence of reduced thermal conductivity on reduced temperature for six niobium samples of different purity. For comparison, a family of theoretical curves [8] for various paramagnetic impurity concentrations have been plotted. One may say that only qualitative agreement exists between the theoretical curves and the experimental data.



## V. SOME ADDITIONAL OBSERVATIONS

Besides the general relationships discussed in the preceeding sections, some additional observations are made in the case of certain superconducting elements for which experimental data are available.

Figure 13 compares, for the case of aluminum, the transition temperatures determined from thermal conductivity data,  $T_c$ , and those determined by electrical measurements,  $T'_c$ . It is noted that transition temperatures determined by thermal conductivity data have a wider range (1.1 to 1.6 K) than transition temperatures determined by electrical method (1.170 to 1.187 K). The same behaviour has been observed for tin but the experimental data are more scattered. In the case of tin, transition temperatures determined from thermal conductivity data are in the range of 3.3 to 3.9 K while those determined by electrical method range from 3.685 to 3.725 K.

Figure 14 shows the dependence of transition temperature of tin on sample purity expressed by the ratio of electrical resistivity. Curve A represents transition temperatures determined from electrical measurements and tends toward an upper limit with increasing sample purity. Curve B corresponds to transition temperatures estimated from thermal conductivity data and does not tend toward a limit (as does curve A) for the most pure samples of tin.

Figure 15 shows the dependence of the impurity parameter  $\beta$  on the purity of the samples of tin expressed by the ratio of electrical resistivity. When presented on logarithmic coordinates one obtains a linear dependence approximated by the equation:

$$\beta = (2.7 \pm 0.1) \times 10^2 \left( \frac{\rho_{273}}{\rho_0} \right)^{-(0.91 \pm 0.01)} \quad (22)$$

To the extent that similar data are not available for other elements, equation (22) can not be tested for general validity.

## VI. DISCUSSIONS AND CONCLUSIONS

As a result of this brief study, two useful relationships have been found, - namely:  
1) the relation between thermal conductivity at the transition temperature and purity of the sample - equation (2), and 2) the relation between transition temperature, purity

of the sample, and thermal conductivity at the transition temperature - equation (6). These two relationships have very simple mathematical forms and physical meaning and are valid to within about 10% for 12 of the 14 superconducting elements investigated. Only indium, mercury, and to some extent lead, do not follow the above mentioned relationships, and it is well known that the theory for mercury and lead is more complicated due to the necessity to take into consideration the so-called "strong coupling effect" [5]. These two general relationships for superconducting elements may be useful because of their simple physical interpretation which may assist in estimating the transition temperature of a pure material from data on impure samples. The observed dependence of the transition temperature on the purity of samples agrees with studies by DeSorbo as quoted in [9] that transition temperature can be decreased by as much as 0.5 K by dissolved oxygen in niobium or increased by a similar amount by strain.

Results presented in this study seem to indicate that for almost all elements investigated transition temperatures based on the thermal conductivity data are higher than transition temperatures reported from electrical measurements. However, these results should be considered as preliminary and further investigations may suggest that higher values of  $T_c$  are related to thermodynamic fluctuations in the metal before creation of Cooper pairs.

The relationships presented in Section 4 are concerned with the dependence of reduced thermal conductivity on reduced temperature for samples of different purity. These relationships have only a qualitative character and indicate a qualitative agreement of the BRT theory and other theories with the experimental data.

In Section 5 some additional observations have been presented. The discrepancy between the transition temperature determined from thermal conductivity data and transition temperatures reported by authors as determined from electrical measurements have been shown. These observations are limited in their conclusiveness due to the lack of more extensive experimental data.

The family of parallel straight lines shown in Figure 2 for twelve elements seem to have a clear physical meaning. Equation (6) which describes these straight lines with the approximation that  $T_c(\beta) = A = \text{constant}$  for each element) may be obtained directly from equation (1) if the term  $\alpha' T^n$  corresponding to the ideal part of thermal resistivity is neglected. In other words, it means that for all these elements the transition temperatures are located on the low temperature branch of the thermal conductivity curve, i. e.  $T_c < T_m$ . Equation (3), which is valid for indium in the case of

impure samples, seems to suggest a little different dependence of the residual electronic thermal resistivity  $W_0$  for the low temperature branch of the thermal conductivity curve for indium; instead of  $W_0 = \beta T^{-1}$  it suggests  $W_0 = \beta^{0.85} T^{-1}$ . One probably may try to explain this difference in the temperature dependence of  $W_0$  for moderately impure indium samples by a concurrent rise in the lattice conduction while the electronic conduction is suppressed by impurities. Equation (4), which is valid for mercury, is obtained for transition temperatures located on the high temperature branch of the thermal conductivity curve, where the term  $\beta T^{-1}$  in equation (1) corresponding to  $W_0$  is small. In Figure 2 the non-linear portion of the curve for lead for a wide range of  $\beta$  probably indicates a comparable contribution of the two parts of thermal resistivities involved in equation (1).

## VII. ACKNOWLEDGMENTS

I would like to express my thanks to the United Nations Development Program for the generous grant which made my extended stay in the United States possible for the period of November 26, 1973 to June 1, 1974. Thanks are also due to the Thermophysical Properties Research Center at Purdue University for receiving me as a guest worker for the period of January 7 to May 16, 1974.

I am indebted to Dr. C. Y. Ho, Assistant Director at TPRC and Head of the Reference Data Division for suggesting the problem and his continued interest and assistance throughout this work. Among the many who were helpful at TPRC, I wish to thank Dr. M. W. Ackerman and Dr. P. E. Liley for many valuable technical discussions. Special thanks are also due to Professors R. J. Sladek of Purdue University, A. C. Anderson at the University of Illinois, J. A. Marcus at Northwestern University, and P. G. Klemens at the University of Connecticut, for many valuable suggestions and contributions.

In concluding, I wish to take this opportunity to express my gratitude to Professor W. Tezciatowski, Director of the Institute for Low Temperature and Structure Research, Polish Academy of Sciences, for making this opportunity available initially and for his concurrence to extending my stay to allow for the conclusion of this study.

## VIII. REFERENCES

1. Ho, C.Y., Powell, R.W., and Liley, P.E., "Thermal Conductivity of the Elements: A Comprehensive Review," Supplement 1 to Volume 3 of the Journal of Physical and Chemical Reference Data, approx. 800 pp., in press.

2. Cezairliyan, A., "Prediction of Thermal Conductivity of Metallic Elements and Their Dilute Alloys at Cryogenic Temperatures," U.S. Air Force Rept. ASD-TDR-63-291, p. 64, 1963.
3. Lynton, E. A., Superconductivity, Third Edition, Spottiswoode, Ballantyne, and Co., Ltd., London and Colchester, p. 101, 1969.
4. Rosenberg, H. G., Low Temperature Solid State Physics, Clarendon Press, Oxford, p. 130, 1963.
5. Ambegaokar, V. and Woo, J., "Calculation of the Thermal Conductivity of Pure Superconducting Lead," Phys. Rev., 139(6A), A1818-23, 1965.
6. Gupta, A. K. and Verma, G. S., "Phenomenological Model for the Electronic Thermal Conductivity of Superconductors," Phys. Rev., 135(1A), A16-9, 1964.
7. Tewordt, L., "Theory of the Intrinsic Electronic Thermal Conductivity of Superconductors," Phys. Rev., 129(2), 657-63, 1963.
8. Ambegaokar, V. and Griffin, A., "Theory of the Thermal Conductivity of Superconducting Alloys with Paramagnetic Impurities," Phys. Rev., 137(4A), A1151-67, 1965.
9. Finnemore, D. K., Stromberg, T. F., and Swenson, C. A., "Superconducting Properties of High-Purity Niobium," Phys. Rev., 149(1), 231-43, 1966.

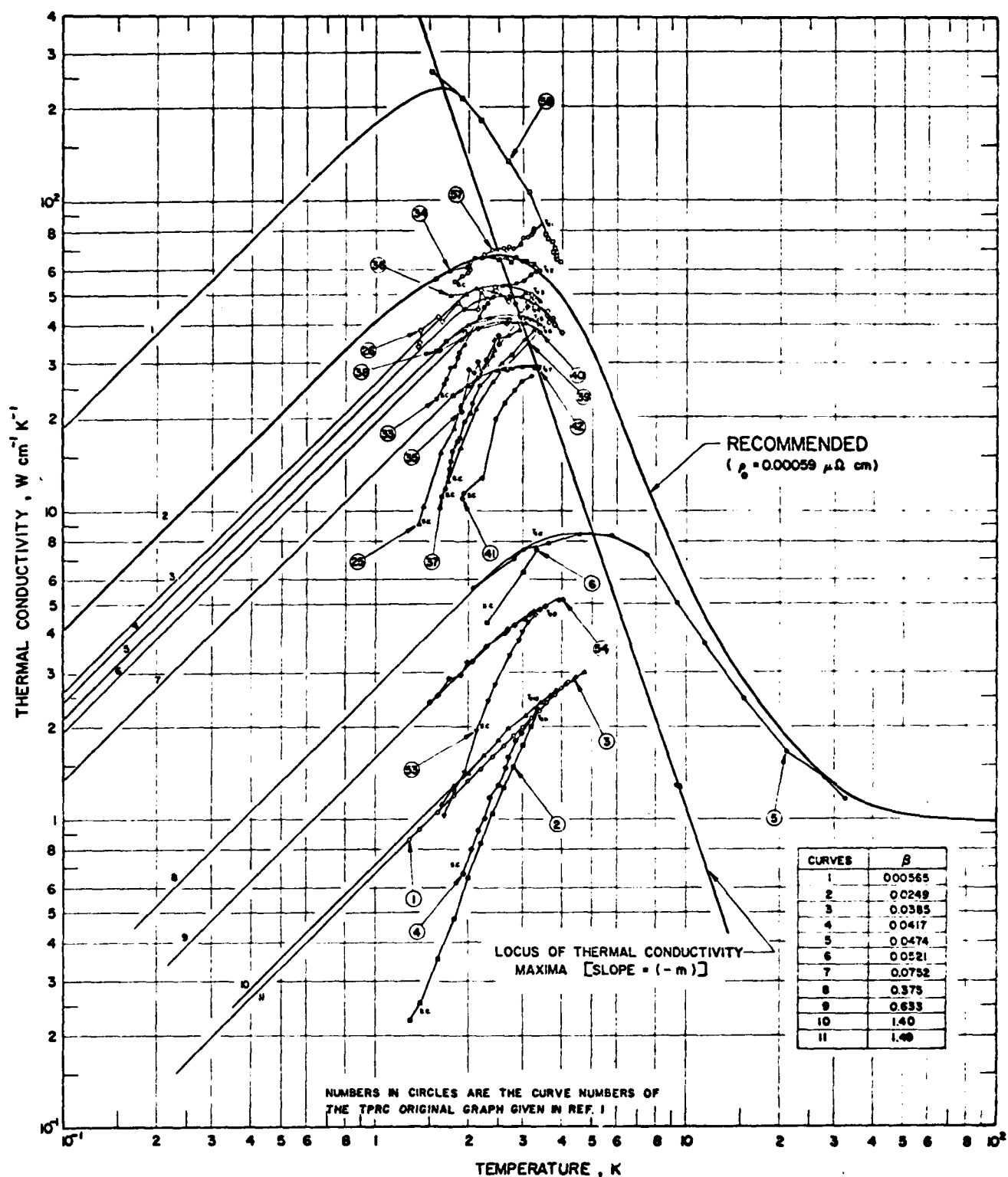


Figure 1. Thermal Conductivity of Indium at Low Temperatures in Normal and Superconducting States.

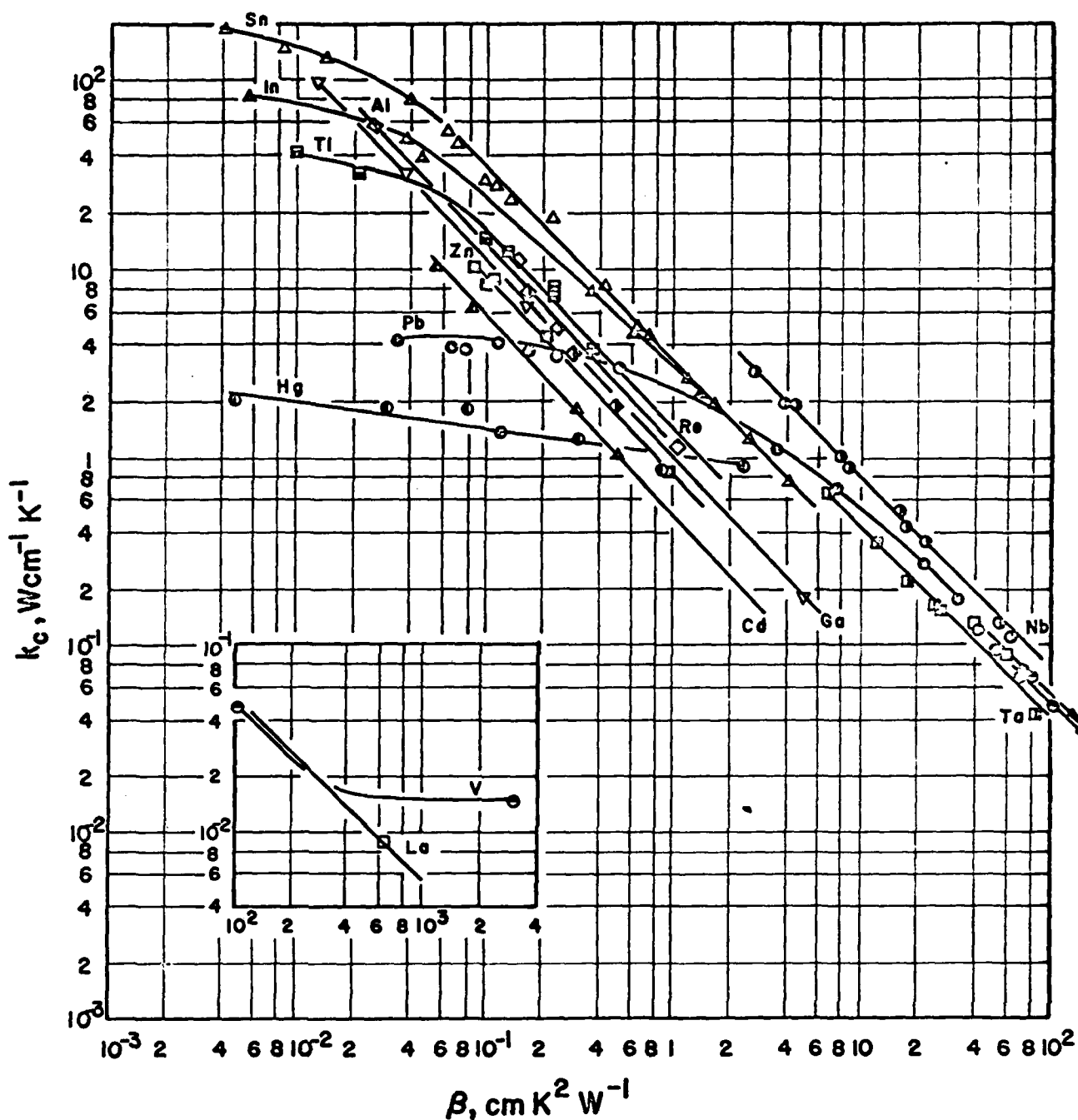


Figure 2. Thermal Conductivity at the Transition Temperature as a Function of Impurity Parameter for 14 Superconducting Elements.

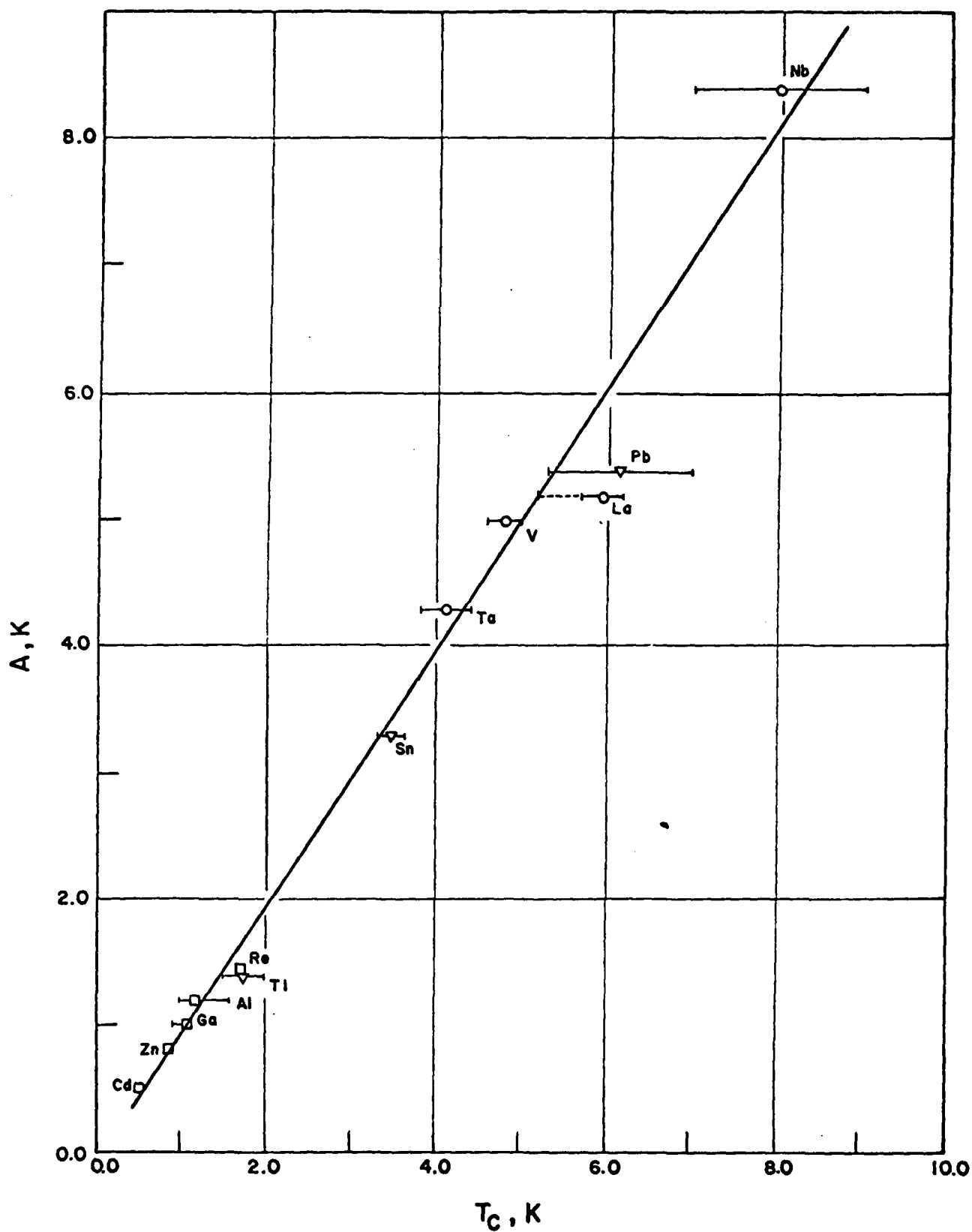


Figure 3. Dependence of the Coefficient  $A$  of Equation:  $k_c = A\beta^{-1}$  on the Transition Temperature for 12 Superconducting Elements.

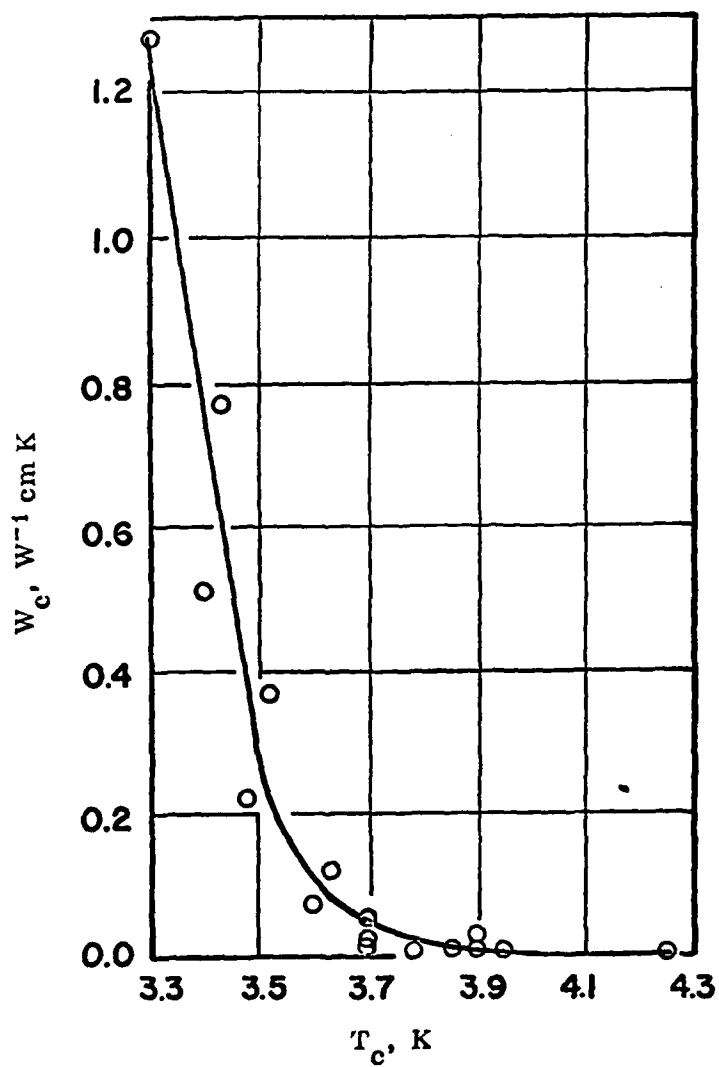


Figure 4. Thermal Resistivity of Tin at Transition Temperature as a Function of the Transition Temperature.



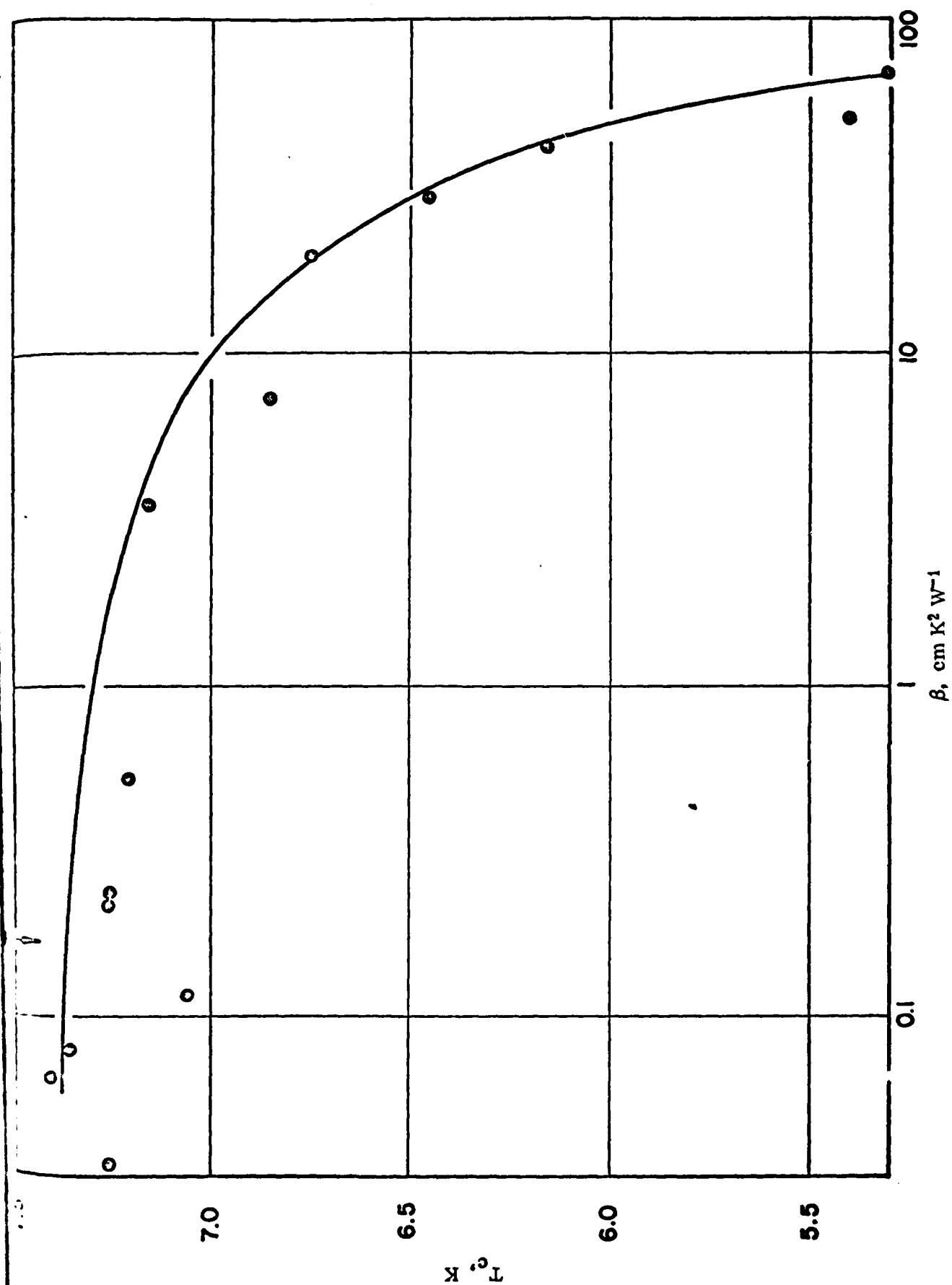


Figure 5. Transition Temperature of Lead as a Function of Impurity Parameter.

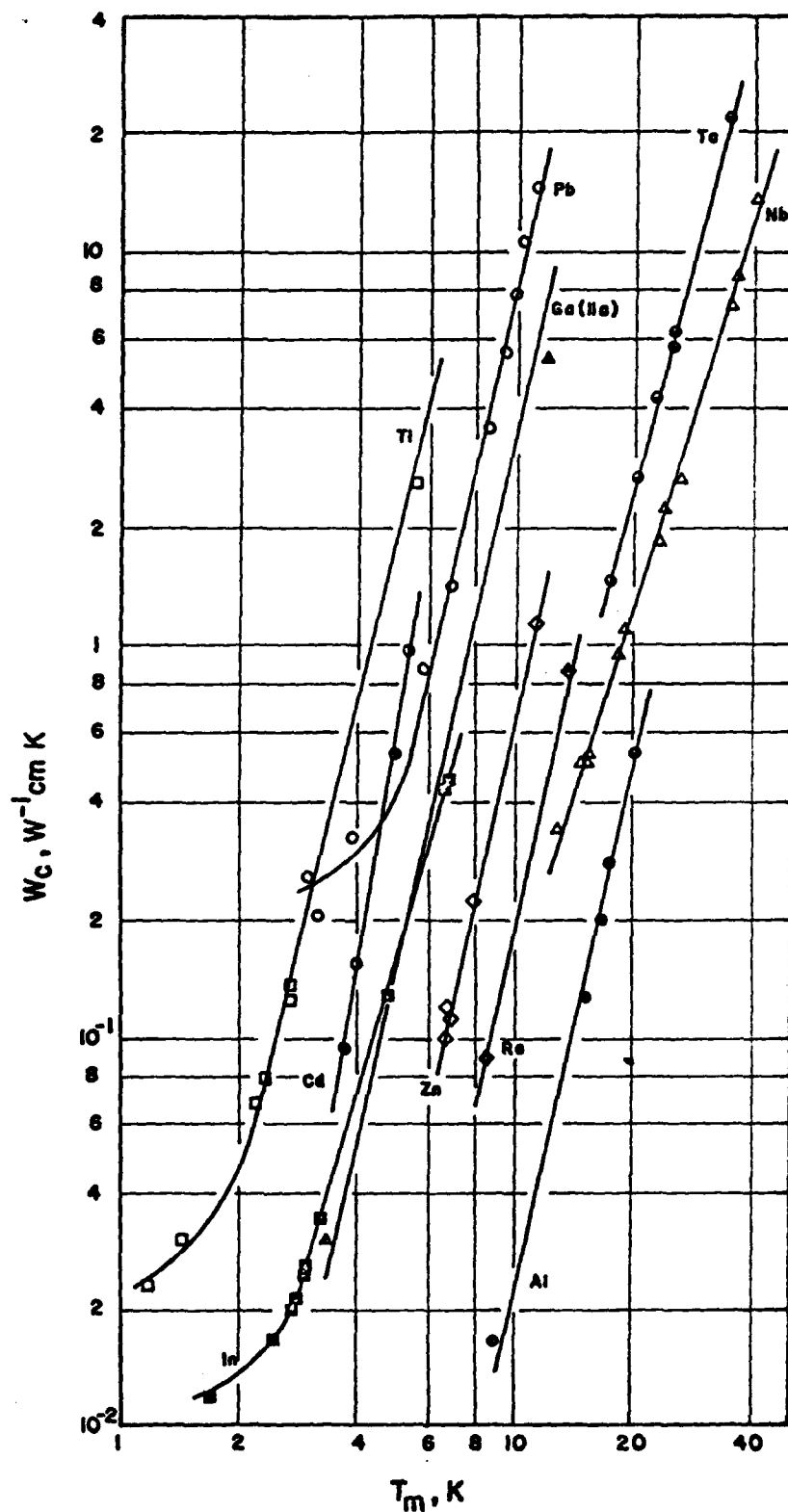


Figure 6. Thermal Resistivity at the Transition Temperature as a Function of Temperature of Thermal Conductivity Maximum for 10 Superconducting Elements.

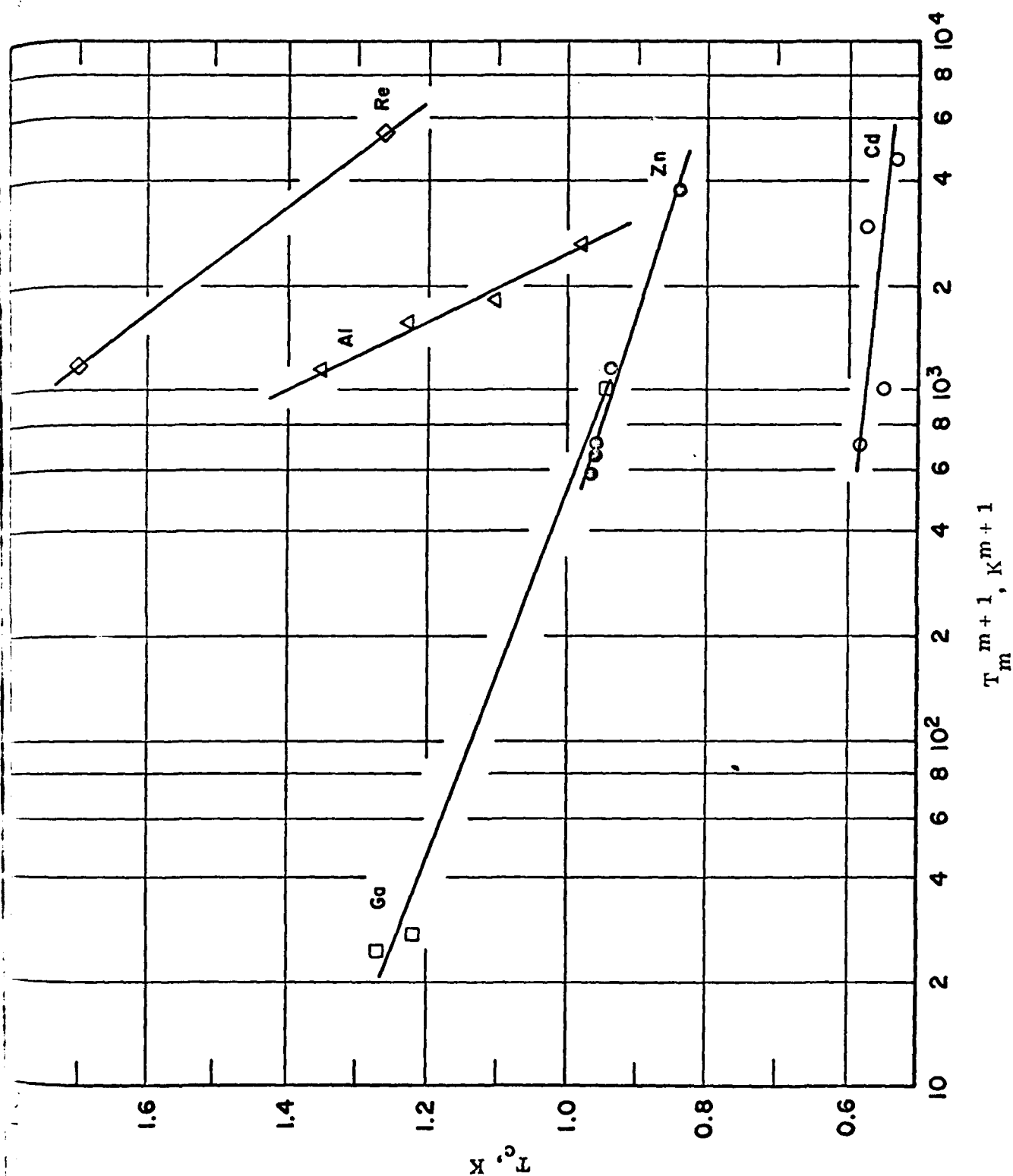


Figure 7. Transition Temperature versus Temperature of Thermal Conductivity Maximum for 5 Superconducting Elements.

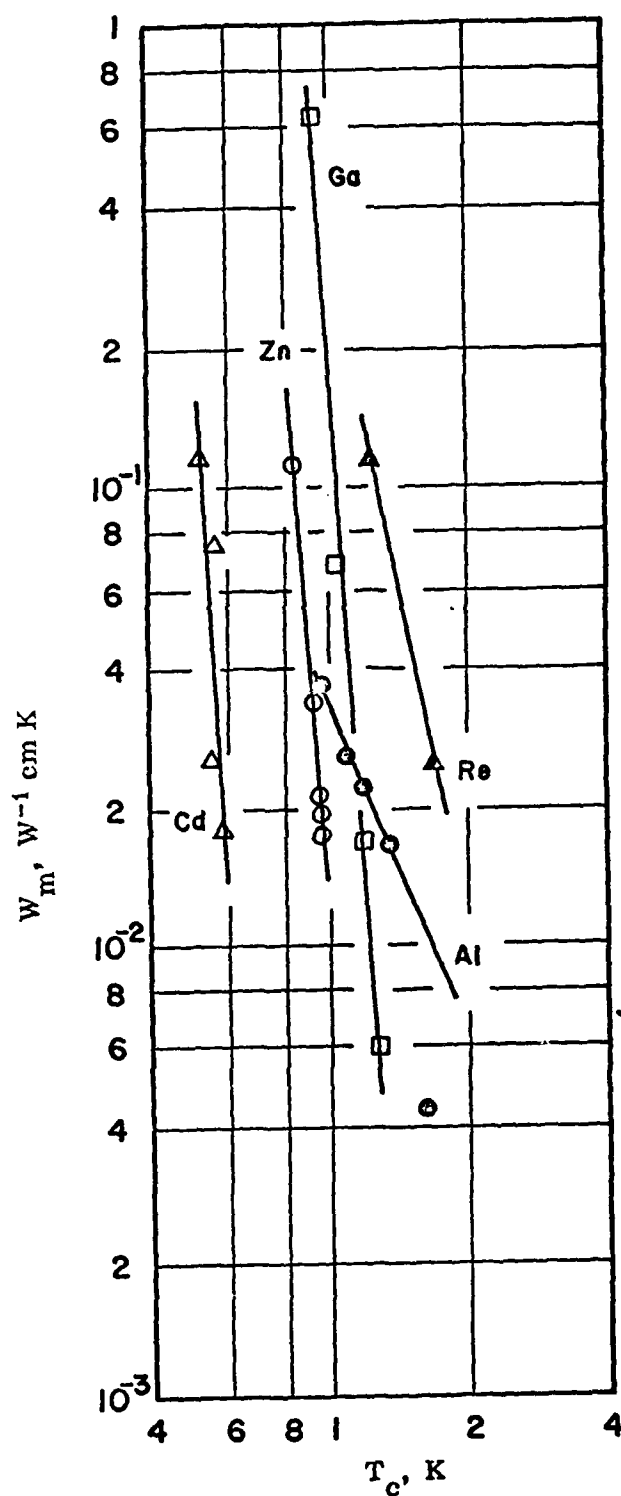


Figure 8. Thermal Resistivity at Temperature of Thermal Conductivity Maximum versus Transition Temperature for 5 Superconducting Elements.

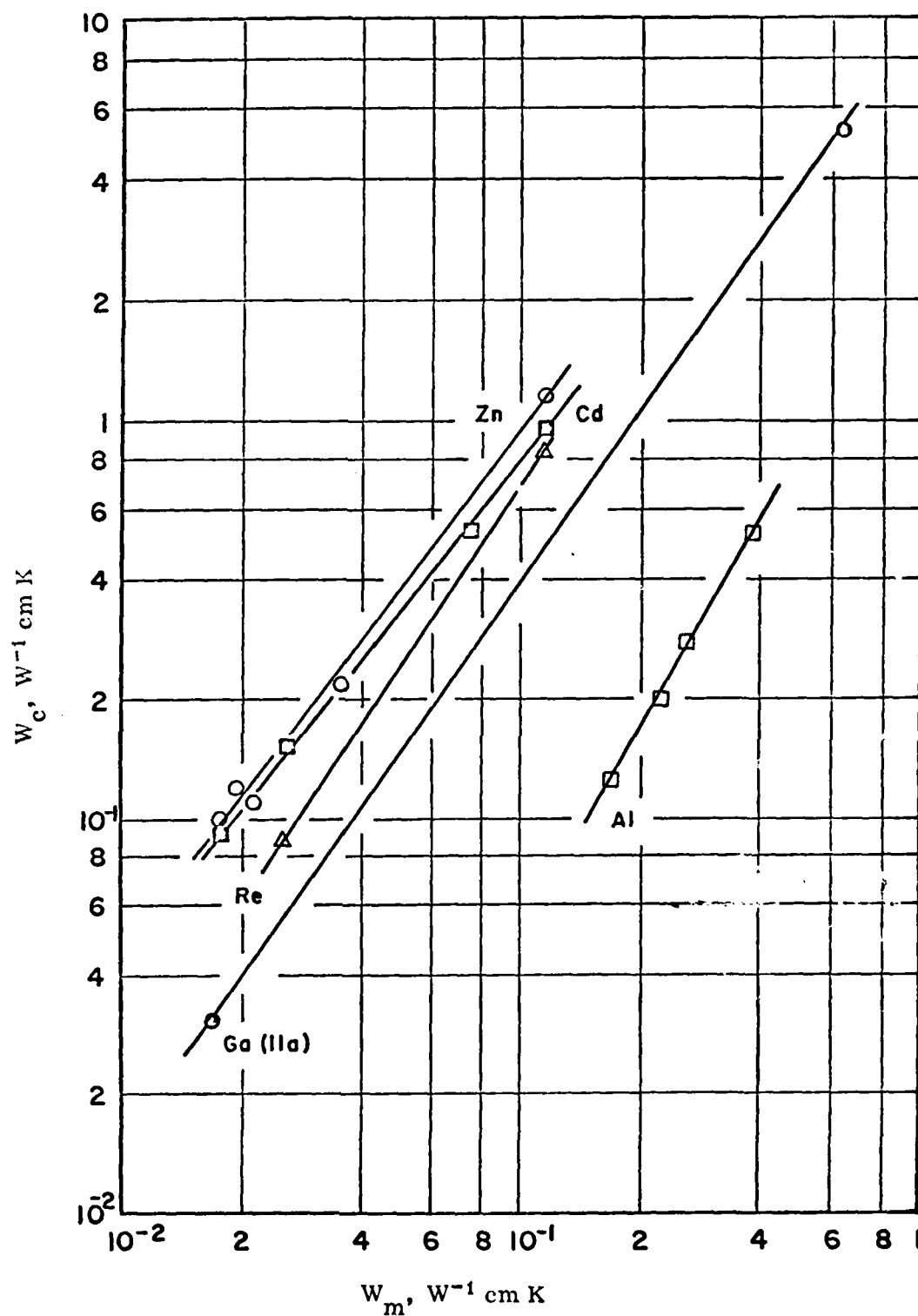


Figure 9. Thermal Resistivity at Transition Temperature versus Thermal Resistivity at Temperature of Thermal Conductivity Maximum for 5 Superconducting Elements.

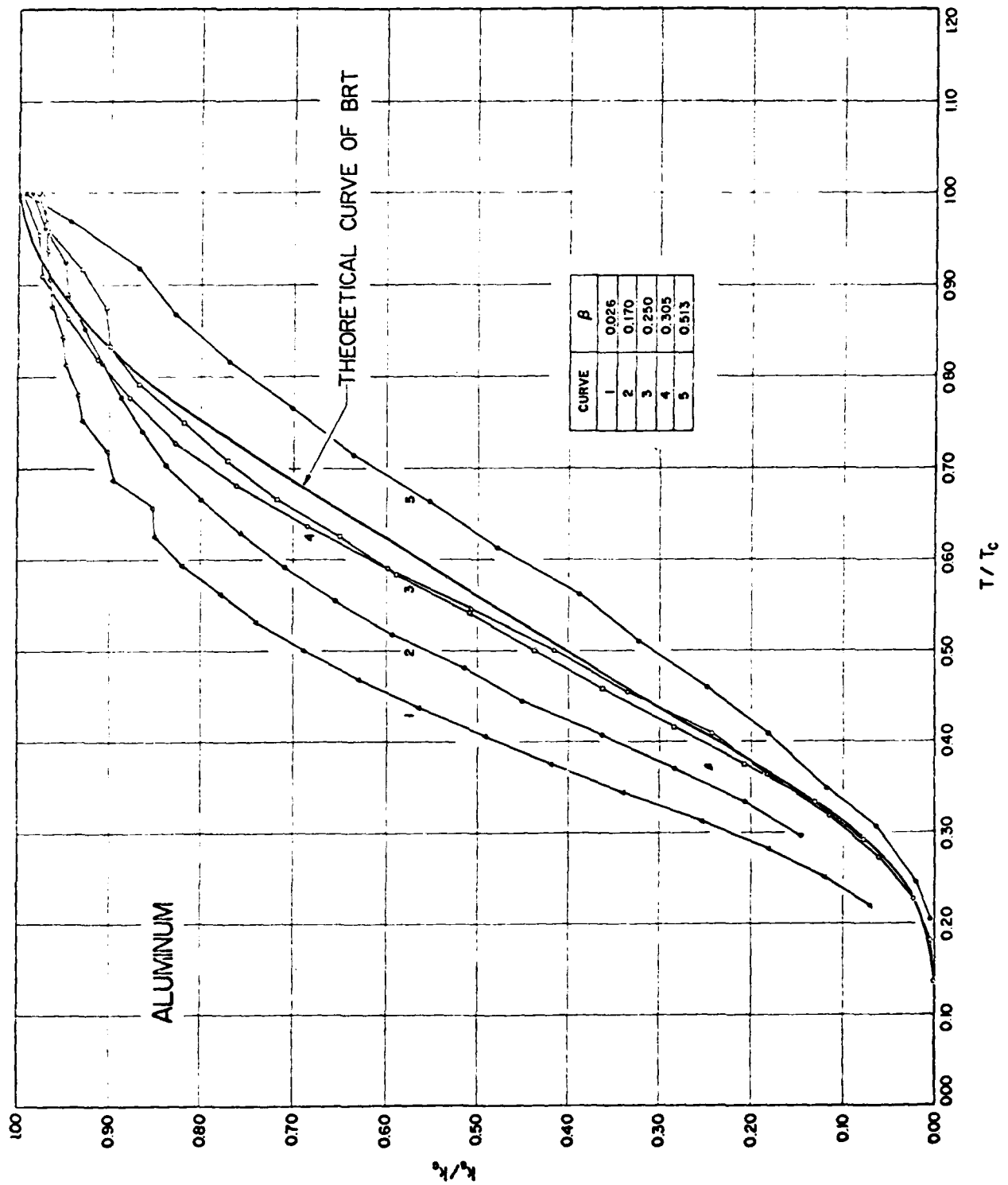


Figure 10. Dependence of Reduced Thermal Conductivity of Aluminum on Reduced Temperature for Samples of Different Purity.

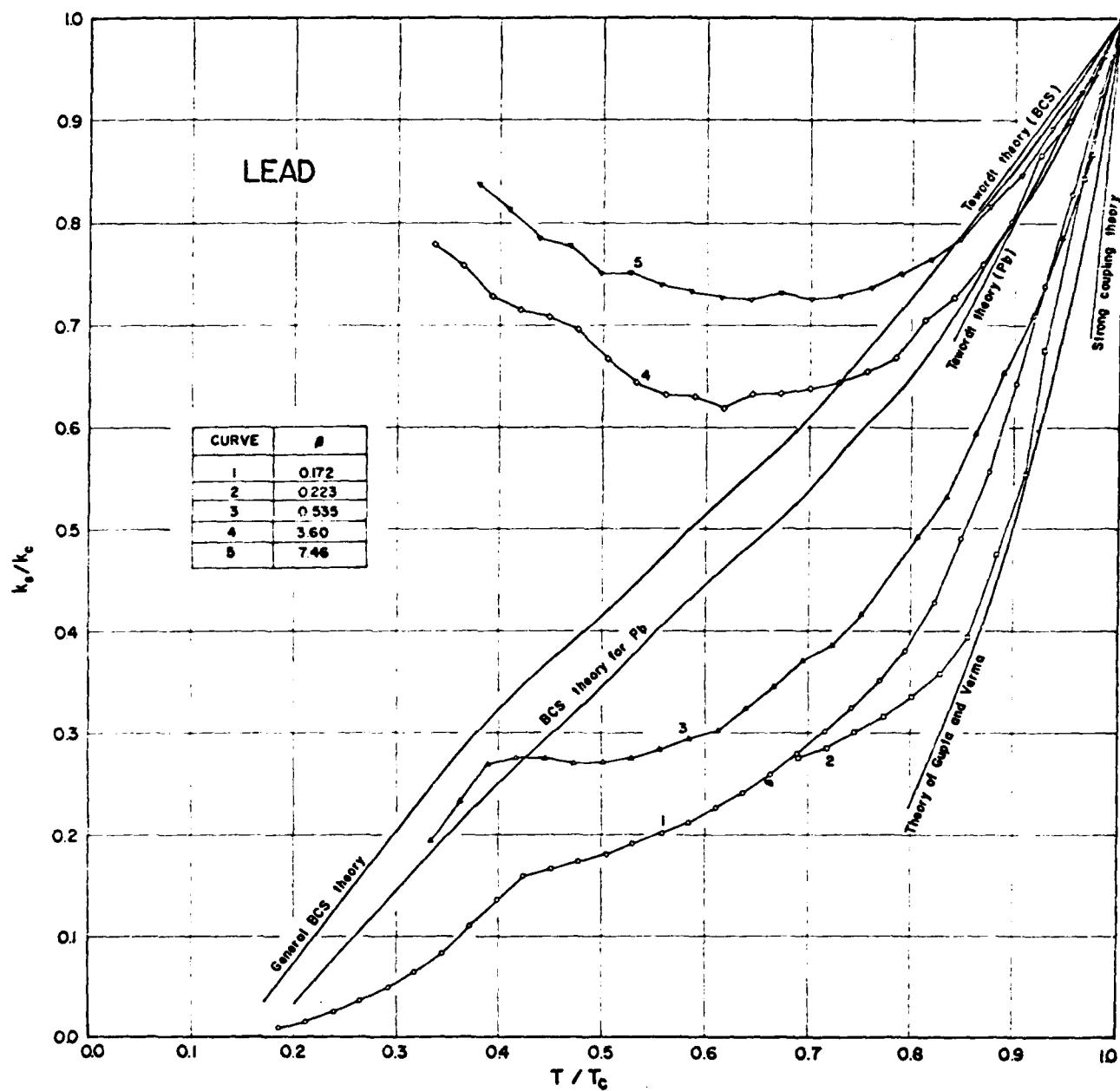


Figure II. Dependence of Reduced Thermal Conductivity of Lead on Reduced Temperature for Samples of Different Purity.

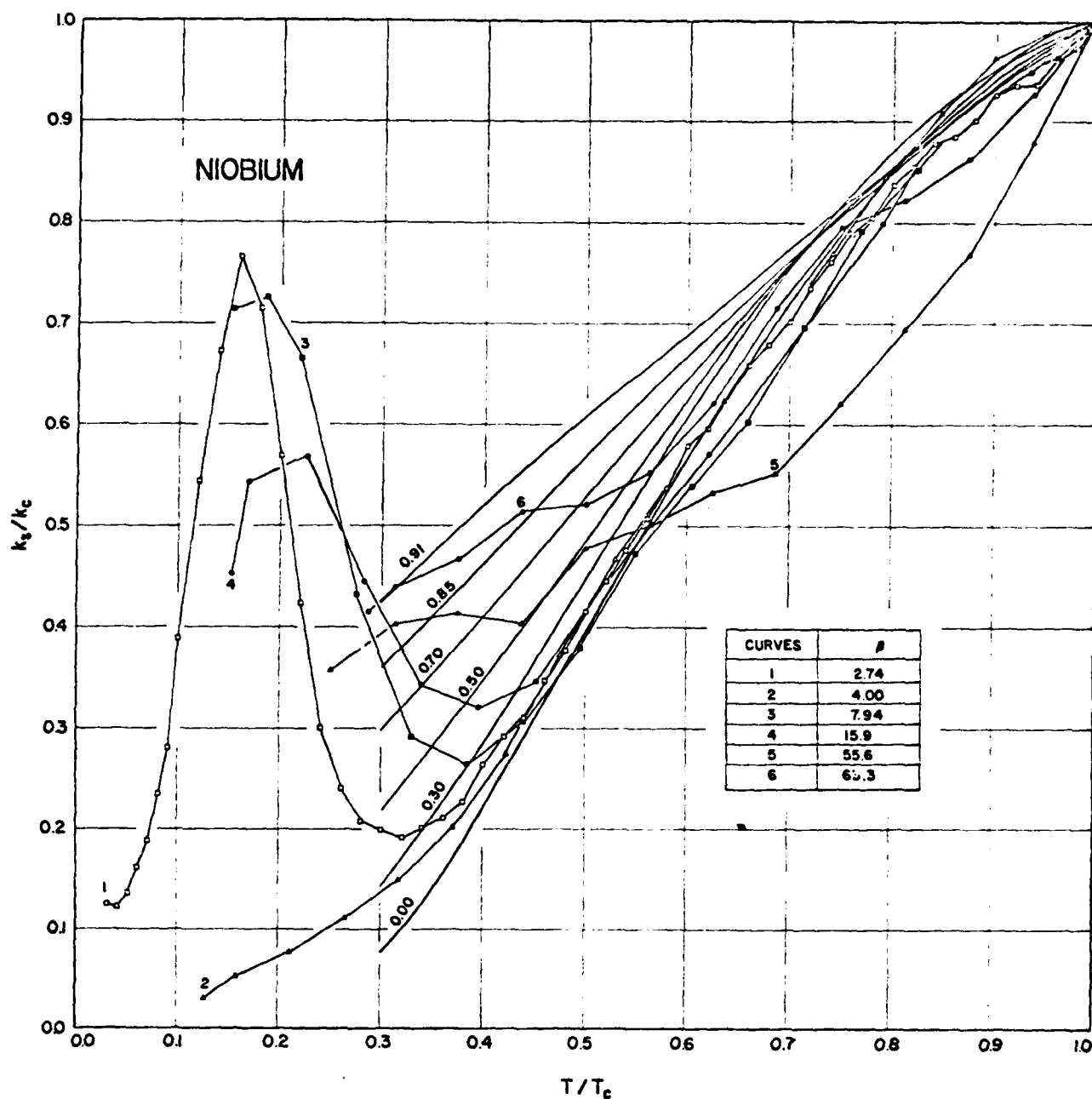


Figure 12. Dependence of Reduced Thermal Conductivity of Niobium on Reduced Temperature For Samples of Different Purity. Curves labeled with numbers 0.00, 0.30, 0.50, 0.70, 0.85, and 0.91 which represent paramagnetic impurity concentrations are theoretical curves based on the model for weak-coupling isotropic superconductor [8].



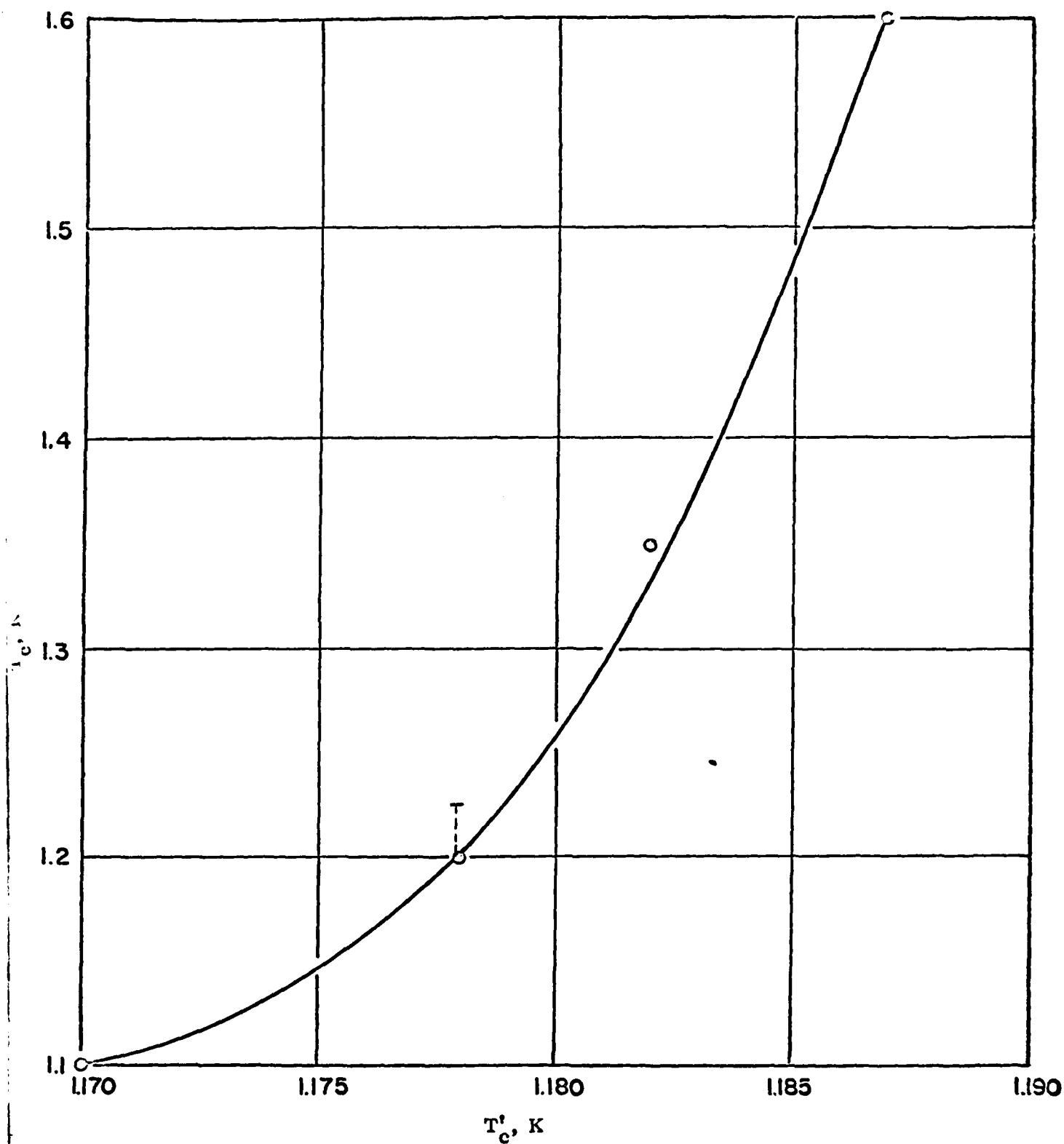


Figure 13. Comparison of Transition Temperatures of Aluminum Samples Determined from Thermal Conductivity Data with those determined from Electrical Measurements.

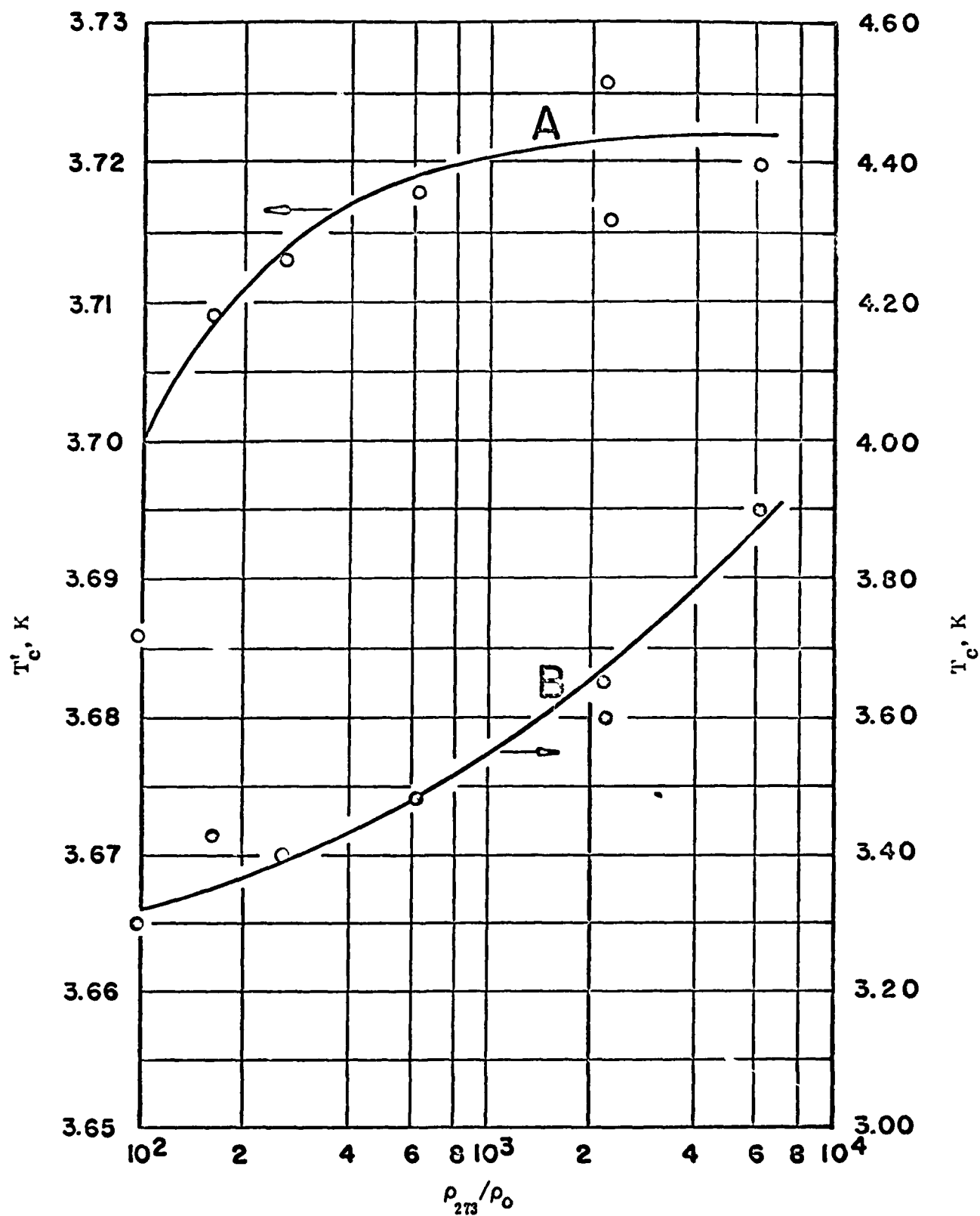


Figure 14. Transition Temperatures of Tin Samples Determined from (A) Electrical Measurements and (B) Thermal Conductivity Data as a Function of Electrical Resistivity Ratio

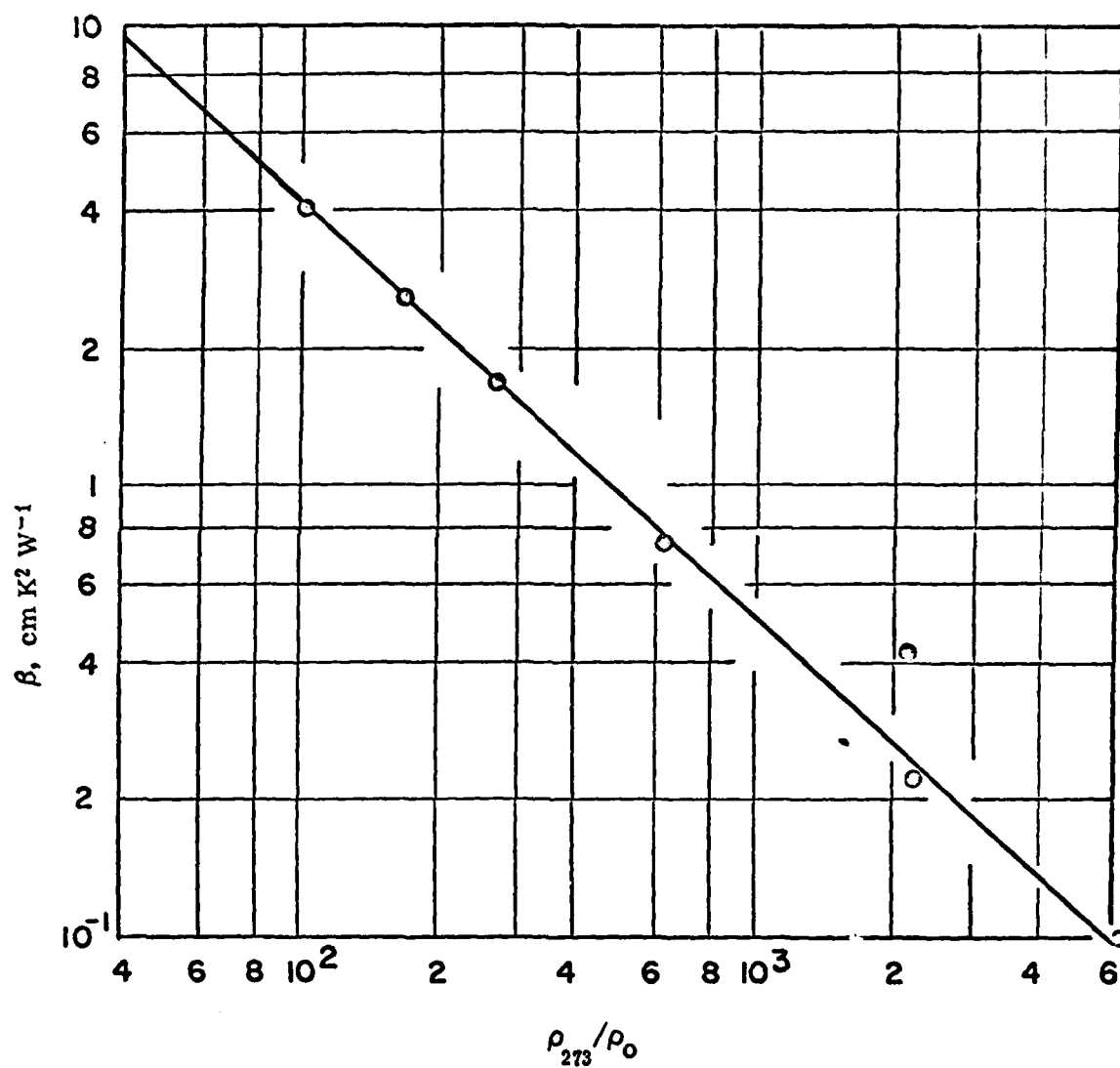


Figure 15. Dependence of the Impurity Parameter on the Electrical Resistivity Ratio for Tin.

Table I. Values of  $T_c$  and  $k_c$  for Samples of Different Purity for all 14 Superconducting Elements Investigated

Element	$T_c$ (K)	$k_c$ (W cm <sup>-1</sup> K <sup>-1</sup> )	$\beta$ (cm K <sup>2</sup> W <sup>-1</sup> )
Elements of the first group			
Al	1.60	60.0	0.086
	1.35	7.85	0.169
	1.225	5.0	0.250
	1.10	3.57	0.305
	0.58	1.50	0.513
Zn	0.965	10.04	0.0862
	0.96	8.4	0.0992
	0.96	9.0	0.111
	0.94	4.5	0.209
	0.84	0.88	0.961
Cd	0.585	10.7	0.0546
	0.550	6.5	0.0847
	0.575	1.87	0.307
	0.533	1.02	0.519
Ga	1.27	98.0	0.0127
	1.22	32.0	0.0374
	1.05	6.38	0.163
	0.94	0.185	5.08
Re	1.698	11.4	0.150
	1.26	1.17	1.07
Elements of the second group			
Pb	7.25	4.35	0.0557
	7.40	4.00	0.0662
	7.35	3.83	0.0816
	7.06	4.26	0.0118
	7.55	3.87	0.172
	7.25	4.95	0.223
	7.25	3.55	0.242
	7.20	3.10	0.535
	7.15	1.15	3.60
	6.85	0.705	7.46
	6.75	0.280	20.2
	6.45	0.178	30.2
	6.15	0.127	42.2
	5.40	0.095	51.6
	5.30	0.069	70.0

Table I (continued)

Element	$T_c$ (K)	$k_c$ (W cm <sup>-1</sup> K <sup>-1</sup> )	$\beta$ (cm K <sup>2</sup> W <sup>-1</sup> )
Elements of the second group (continued)			
In	3.46	83.2	0.00565
	3.40	60.0	0.0249
	3.375	49.2	0.0385
	3.30	47.2	0.0417
	3.35	40.05	0.0474
	3.35	38.5	0.0521
	3.28	29.5	0.0752
	3.35	7.9	0.0374
	3.30	4.75	0.633
	3.35	2.30	1.40
	3.30	2.20	1.49
Sn	4.25	190.0	0.00422
	3.95	150.0	0.00858
	3.90	146.0	0.00935
	3.85	134.0	0.0144
	3.78	81.5	0.0405
	3.70	76.5	0.0435
	3.50	54.8	0.0629
	3.70	47.5	0.0704
	3.90	30.5	0.102
	3.50	28.75	0.115
	3.48	23.80	0.133
	3.45	19.75	0.167
	3.70	19.7	0.172
	3.60	13.8	0.229
	3.65	8.4	0.431
	3.25	4.85	0.656
	3.48	4.5	0.749
	3.52	2.72	1.20
	3.40	1.97	1.69
	3.43	1.30	2.61
	3.30	0.785	4.17
Hg	4.25	2.05	0.00476
	4.20	1.85	0.0254
	4.20	1.85	0.0294
	4.20	1.85	0.0806
	4.20	1.30	0.117
	4.15	1.40	0.117
	4.10	1.30	0.312
	4.10	0.87	0.877
	4.05	0.92	2.41

Table I (continued)

Element	$T_c$ (K)	$k_c$ (W cm <sup>-1</sup> K <sup>-1</sup> )	$\beta$ (cm K <sup>2</sup> W <sup>-1</sup> )
Elements of the second group (continued)			
Tl	2.62	43.3	0.00997
	2.75	32.8	0.0208
	2.68	14.7	0.108
	2.70	12.75	0.132
	2.37-1.95	8.1-7.4	0.230
	1.54	0.38	3.73
Elements of the third group			
Nb	10.0	2.93	2.74
	9.45	2.0	4.00
	9.40	1.95	4.44
	9.50	1.96	4.44
	9.60	1.90	4.69
	9.05	1.05	7.94
	8.90	0.91	8.85
	8.80	0.54	15.9
	8.00	0.44	17.4
	8.50	0.37	22.7
	8.00	0.135	55.5
	8.00	0.115	63.3
	7.00	0.072	88.9
Ta	4.70	0.665	7.02
	4.50	0.375	12.3
	4.40	0.232	18.2
	4.35	0.172	25.0
	4.25	0.160	24.7
	3.85	0.045	84.8
V	5.45	0.0755	70.42
	5.80	0.070	81.63
	4.95	0.047	104.7
	4.60	0.0152	3030
La	6.80	0.137	40.0
	6.20	0.0920	59.2
	5.70	0.0089	649

Table II. Values of Coefficient A of equation:  
 $k_c = A\beta^{-1}$  for 12 Superconducting Elements

<u>Element</u>	<u>A (K)</u>
Al	$1.20 \pm 0.04$
Zn	$0.79 \pm 0.1$
Cd	$0.50 \pm 0.06$
Ga	$1.0 \pm 0.1$
Re	$1.45 \pm 0.03$
Pb	$5.4 \pm 0.2$
Sn	$3.3 \pm 0.2$
Tl	$1.4 \pm 0.1$
Nb	$8.40 \pm 0.15$
Ta	$4.3 \pm 0.1$
V	$5.0 \pm 0.3$
La	$5.2 \pm 0.4$

Table III. Values of coefficients C and D of Equation (7) for 13 Superconducting Elements

<u>Element</u>	<u>C (cm K W<sup>-1</sup>)</u>	<u>D (cm W<sup>-1</sup>)</u>
Al	0.72 ± 0.04	0.45 ± 0.03
Zn	8.6 ± 0.3	9.2 ± 0.3
Cd	26.7 ± 0.2	48.7 ± 0.4
Ga	47.6 ± 0.7	45.7 ± 0.7
Pb	50.5 ± 0.25	6.98 ± 0.03
In	21.8 ± 0.1	6.50 ± 0.04
Sn	17.7 ± 0.3	5.1 ± 0.16
Hg	15.3 ± 0.6	3.7 ± 0.2
Tl	11.7 ± 0.4	6.2 ± 0.3
Nb	58 ± 3	6.7 ± 0.3
Ta	144 ± 4	32.7 ± 1
V	610 ± 30	125.0 ± 6
La	1240 ± 35	203 ± 5



Table IV. Ranges of  $\beta$  and  $T_c$  in which  
Equations (2, 6-10) Are Valid

Element	Range of $\beta$ where $k_c = \frac{A}{\beta}$	Range of $T_c$ where $k_c = (C - DT_c)^{-1}$	Range of $\beta$ corresponding to the range of $T_c$
Al	0.025-0.25	1.0-1.6	0.026-0.45
Zn	0.086-1.0	0.84-0.94	0.21-0.95
Cd	0.05-0.5	0.533-0.55	0.5-0.54
Ga	0.01-5.0	0.84-1.04	0.82-5.0
Re	0.15-1.1	-	-
Pb	7.0-70.0	5.30-7.20	2.8-68.0
In	-	3.30-3.36	0.017-0.13
Sn	0.04-4.5	3.30-3.35	0.68-5.6
Hg	-	4.05-4.20	0.08-2.25
Tl	0.08-4.0	1.55-1.90	1.9-3.75
Nb	3.0-90.0	7.0-8.8	15-68
Ta	7-85	3.85-4.40	18-85
V	70-3000	4.6-4.9	13-1500
La	40-650	5.7-6.2	135-650

Table V. Calculated values of Thermal Conductivity Maximum and Its Corresponding Temperature for Different Values of the Impurity Parameter  $\beta$  for 10 Superconducting Elements

Element	$\beta$ (cm K <sup>2</sup> W <sup>-1</sup> )	$T_m$ (K)	$k_m$ (W cm <sup>-1</sup> K <sup>-1</sup> )
Al	0.026	8.882	227.7
	0.170	14.920	58.51
	0.250	16.598	44.26
	0.305	17.535	38.33
	0.513	20.243	26.31
Zn	0.0525	5.827	83.24
	0.0862	6.522	56.75
	0.0992	6.734	50.91
	0.111	6.909	46.64
	0.209	7.575	28.64
	0.961	11.283	8.80
Cd	0.00456	2.450	439.7
	0.0546	3.707	55.51
	0.0847	3.988	38.51
	0.307	4.942	13.18
	0.519	5.395	8.50
Ga	0.00409	1.840	299.9
	parallel to a axis	0.0374	3.306
	parallel to b axis	5.08	12.114
	parallel to c axis	0.0014	1.771
		0.0127	3.177
		0.0174	1.931
Re	0.150	8.593	39.38
	1.07	13.585	8.68
Pb	0.172	2.996	13.03
	0.223	3.173	10.66
	0.535	3.853	5.40
	3.60	5.885	1.22
	7.46	6.921	0.686
	20.2	8.636	0.321
	30.3	9.449	0.234
	42.2	10.170	0.181
	51.6	10.636	0.155
	70.0	11.382	0.122
In	0.00565	1.695	188.9
	0.0249	2.442	65.39
	0.0385	2.723	47.16
	0.0417	2.778	44.41
	0.0474	2.869	40.35
	0.0521	2.937	37.58
	0.0752	3.219	28.54
	0.374	4.809	8.56
	0.633	5.483	5.78
	1.40	6.686	3.19
	1.49	6.783	3.03

Table V (continued)

<u>Element</u>	<u><math>\beta</math> (cm K<sup>2</sup> W<sup>-1</sup>)</u>	<u>T<sub>m</sub> (K)</u>	<u>k<sub>m</sub> (W cm<sup>-1</sup> K<sup>-1</sup>)</u>
Tl	0.00982	1.180	80.09
	0.00997	1.184	79.20
	0.0208	1.438	46.02
	0.108	2.218	13.68
	0.132	2.335	11.83
	0.230	2.705	7.84
	3.73	5.632	1.01
Nb	2.74	13.017	3.17
	4.00	14.768	2.46
	4.44	15.296	2.29
	4.69	15.578	2.21
	7.94	18.557	1.56
	8.85	19.243	1.45
	15.9	23.380	0.982
	17.4	24.103	0.924
	22.7	26.352	0.773
	55.5	35.498	0.426
	63.3	37.074	0.380
	88.9	41.518	0.311
Ta	7.02	17.520	1.66
	12.3	20.552	1.11
	18.2	22.950	0.838
	25.0	25.084	0.669
	26.7	25.546	0.639
	84.7	35.414	0.278

Table VI. Values of coefficient E of Equation (11)  
for 6 Superconducting Elements

<u>Element</u>	<u>E (cm W<sup>-1</sup> K<sup>-3</sup>)</u>
Al	$(0.90 \pm 0.05) \times 10^{-6}$
Zn	$(2.4 \pm 0.2) \times 10^{-5}$
Ga (parallel to a axis)	$(1.3 \pm 0.3) \times 10^{-4}$
Re	$(8 \pm 1) \times 10^{-6}$
Pb	$(3.0 \pm 0.3) \times 10^{-4}$
Tl	$(18 \pm 2) \times 10^{-4}$

END

DATE  
FILMED

6-83

DTIC